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EFFECTIVE MEDIUM THEORIES OF
INHOMOGENEOUS MEDIA FROM A MODERN PERSPECTIVE

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INTRODUCTION

Many physical phenomena are associated with inhomogeneous medium. For example, a solid state physicist may study the electrical properties of disordered alloys; a geologist, the mechanical properties of polycrystalline rocks. Other scientists may ponder the twinkling of stars or water-seepage through concrete.

Various theoretical approaches to these diverse phenomena generally exhibit two common features. One feature is a statement of physics, usually in the context of a model. This statement may be the equation of motion for a solid continuum, the time independent Schrödinger equation, Helmholtz's equation, etc. These various equations regulate the dynamical variables and have as parameters stochastic variables associated with the inhomogeneous medium. The second common feature is an average of the dynamical variables or their products over the distribution of the stochastic parameters. Even when the statistical information is complete, only an approximation to the averaging, which for some problems is called an effective medium approximation, is generally possible. Most often effective medium approximations are developed by intuitive means.

Recently, in solid state physics in the study of disordered alloys, theorists stated the physics of their problem in terms of an integral equation and analyzed this equation by techniques developed in the quantum mechanical theory of scattering. This integral equation is equivalent to a perturbation series (an infinite series), and various effective medium approximations were developed usually by approximating the average of each term in the series and then summing an infinite series of terms. This approach, scattering theory with infinite order perturbation summation, developed perturbatively several effective medium approximations. Some of these approximations have direct analogs to approximations developed intuitively for other phenomena.

I will illustrate the application of the scattering theory approach to a problem outside of solid state physics, the computation of the effective dielectric constant of a polycrystal. I will state the problem in the form of an integral equation, recover several well-known intuitive approximations, and indicate, but not demonstrate, the connection of the approximations to perturbation theory. This example and its discussion is intended primarily as an illustration of an approach to problems associated with inhomogeneous media that has the advantages of being formally general, having a specific recipe for its application, and having the potential for error analysis. Additionally, the discussion of the approximations hopefully clarifies the meaning of several commonly used effective medium approximations.

Because of an apparent growing interest to use the scattering theory approach in scientific areas other than solid state physics, part of the discussion is quite detailed to provide an introduction to concepts and structures which may appear new. The other part of the discussion is

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considerably less detailed. In place of detail are merely remarks about the nature of these commonly used approximations. In some cases considerable analysis is required for a full appreciation of these remarks; some references to literature where the analysis is detailed are given.

SCATTERING THEORY APPROACH

Statement of the Problem

The example under consideration is a polycrystalline material that has a dielectric constant tensor $\underline{\epsilon}(\underline{r})$ which in general changes from grain to grain because of changes in material type, orientation of the grain, or grain size and shape. However, when measured, the material as a whole behaves effectively as a homogeneous medium with a dielectric constant $\underline{\epsilon}^*$. The problem is to calculate $\underline{\epsilon}^*$ from statistical information about $\underline{\epsilon}(\underline{r})$.

For a static problem the physics follows from

$$\nabla \cdot \underline{D} = 0 \quad (1)$$

where \underline{D} is the electric displacement field. Modelling of the medium begins with the constitutive relation

$$\underline{D}(\underline{r}) = \underline{\epsilon}(\underline{r}) \underline{E}(\underline{r}) \quad (2)$$

where \underline{E} is the electric field intensity, and the effective dielectric constant is defined by

$$\langle \underline{D} \rangle = \underline{\epsilon}^* \langle \underline{E} \rangle \quad (3)$$

where the angular brackets denote ensemble averaging.

The problem specified by Eqs. (1)-(3) is analogous to many other problems. As \underline{E} is a gradient of a potential, replacing \underline{E} by the gradient of temperature and \underline{D} by the heat current re-interprets $\underline{\epsilon}$ as a thermal conductivity. Through similar re-interpretations the problems of effective electrical conductivity, permeability, diffusivity, elastic stiffness, etc. are seen as directly analogous. The problem of effective elastic properties, of course, requires increases in tensorial rank.

It is useful to write $\underline{\epsilon}$ as the sum of two parts

$$\underline{\epsilon} = \underline{\epsilon}^0 + \underline{\delta\epsilon} \quad (4)$$

where $\underline{\epsilon}^0$ is some arbitrarily, but in general conveniently, chosen spatially invariant (homogeneous) dielectric constant so that all stochastic variations are contained in the perturbation $\underline{\delta\epsilon}$. One can now show the equivalence of Eqs. (1)-(4) to the integral equation

$$\underline{E} = \underline{E}^0 + \int d\underline{r}' [\underline{\delta\epsilon}(\underline{r}') \underline{E}(\underline{r}') \cdot \nabla'] \underline{g}(\underline{r}, \underline{r}') \quad (5)$$

where \underline{g} is the Green's function satisfying

$$\nabla \cdot \underline{\epsilon}^0 \underline{g} = \delta(\underline{r} - \underline{r}')$$

and \underline{E}^0 satisfies the homogeneous equation

$$\nabla \cdot \underline{\epsilon}^0 \underline{E}^0 = 0$$

It is also useful to write the integral equation in several different forms. First, with a standard indicial notation

$$E_i = E_i^0 + \int d\underline{r}' G_{ij} \delta\epsilon_{jk} E_j$$

where

$$G_{ij} = G_{ji} = g_{i,j} = \phi_{,ij}$$

since g_i is expressible as the gradient of a potential ϕ . Next, in an operator notation

$$E = E^0 + G\delta\epsilon E \quad (6)$$

where G is an integral operator

$$Gf \rightarrow \int d\underline{r}' G_{ij}(\underline{r}, \underline{r}') f_j(\underline{r}')$$

Equation (6) is a statement of the physics that is identical to Eqs. (1)-(3). It is analogous to the Lippman-Schwinger (the scattering) equation in quantum mechanics.

The integral equation has a formal solution obtained by iteration and represented by the infinite series

$$E = E^0 + G\delta\epsilon E^0 + G\delta\epsilon G\delta\epsilon E^0 + \dots$$

Different terms contain the perturbation $\delta\epsilon$ to different "powers". With the definition of T , called the T -matrix in quantum mechanics,

$$T = \delta\epsilon + \delta\epsilon G \delta\epsilon + \delta\epsilon G \delta\epsilon G \delta\epsilon + \dots$$

the following equivalent to (6) is obtained

$$E = E^0 + GTE^0 \quad (7)$$

Now T represents the perturbation series, and a formal summation of T is obvious

$$T = \delta\epsilon (I - G\delta\epsilon)^{-1}$$

Since the explicit character of $\delta\epsilon$ is yet unspecified, Eqs. (6) and (7) are general. The specification of $\delta\epsilon$ for a polycrystal assumes the following model:

$$\delta\epsilon(\underline{r}) = \sum_{\alpha} \delta\epsilon^{\alpha} \Theta^{\alpha}(\underline{r}) \quad (8)$$

where $\Theta^{\alpha}(\underline{r}) = 1, \underline{r}$ in grain α
0, otherwise

The term "grain" is used in the broadest possible sense. For example, the region α could be a pore.

To correspond to the piecewise behavior of $\delta\epsilon$, it is convenient to define a t -matrix associated with grain α

$$t^{\alpha} = \delta\epsilon^{\alpha} \Theta^{\alpha} (I - G\delta\epsilon^{\alpha} \Theta^{\alpha})^{-1} \quad (9)$$

With this definition T is expressible as

$$T = \sum_{\alpha} t^{\alpha} + \sum_{\alpha} \sum_{\beta \neq \alpha} t^{\alpha} G t^{\beta} + \sum_{\alpha} \sum_{\beta \neq \alpha} \sum_{\gamma \neq \beta} t^{\alpha} G t^{\beta} G t^{\gamma} + \dots \quad (10)$$

Although (10) is equivalent to (7), it shifts the focus of the perturbation series from $\delta\epsilon$ to t^{α} . The significance is that if (7) were truncated after the first term, the perturbation series would be first order in $\delta\epsilon^{\alpha}$. On the other hand, if (10) is truncated with the first term, the perturbation would be of infinite order in $\delta\epsilon^{\alpha}$.

t^{α} has a physical meaning: If the deviation from homogeneity is confined solely to one region α , then

$$E = E^{\circ} + G \delta\epsilon^{\alpha} \Theta^{\alpha} E$$

which when iterated and summed becomes

$$E = E^{\circ} + G t^{\alpha} E^{\circ}$$

Thus t^{α} is the T-matrix which solves the single inhomogeneity problem. Consequently in (10) the first term represents contributions from regions α individually embedded in a homogeneous medium ϵ° . The remaining terms, all involving at least two regions, represent the interaction between regions.

The Averaging

To compute ϵ^* , $\langle E \rangle$ and $\langle D \rangle$ are needed. $\langle E \rangle$ is determined directly from (7)

$$\langle E \rangle = E^{\circ} + \langle GT \rangle E^{\circ} \quad (11)$$

but the determination of $\langle D \rangle$ involves several steps. First, since $\epsilon = \epsilon^{\circ} + \delta\epsilon$,

$$D = \epsilon^{\circ} E + \delta\epsilon E$$

Next from a comparison of (6) and (7),

$$\delta\epsilon E = T E^{\circ}$$

Finally,

$$\langle D \rangle = \epsilon^{\circ} \langle E \rangle + \langle T \rangle E^{\circ} \quad (12)$$

Thus, (11) and (12) with (3) yield

$$\epsilon^* = \epsilon^{\circ} + \langle T \rangle (I + \langle GT \rangle)^{-1} \quad (13)$$

This is an exact equation, independent of the assumed polycrystalline model.

As with most exact equations, exact evaluation is usually impossible. Approximations are needed. In the present case, one sees that approximations to T are especially important, and one possible approximation is to truncate (10) after the first term.

$$T \approx \sum_{\alpha} t^{\alpha} \quad (14)$$

For the standard problem of isotropic $\delta\epsilon^{\alpha}$ and spherical grains, the above when used in (13) yields

$$\frac{\epsilon^* - \epsilon^{\circ}}{\epsilon^* + 2\epsilon^{\circ}} = \sum_j f_j \frac{\epsilon^j - \epsilon^{\circ}}{\epsilon^j + 2\epsilon^{\circ}} \quad (15)$$

where f_j is the volume fraction of material type j that has a dielectric constant ϵ^j . Often, a polarizability

$$\frac{4\pi\alpha^j}{3} = \frac{\epsilon^j - \epsilon^{\circ}}{\epsilon^j + 2\epsilon^{\circ}}$$

is associated with each grain, and (15) is written as

$$\frac{\epsilon^* - \epsilon^{\circ}}{\epsilon^* + 2\epsilon^{\circ}} = \frac{4\pi}{3} \sum_j f_j \alpha^j \quad (16)$$

This is the famous Clausius-Mossotti equation or Lorentz-Lorenz formula.

It is important to note that (14) is used in (13), not in

$$\epsilon^* = \epsilon^{\circ} + \langle T \rangle \quad (17)$$

Since t^{α} solves the problem of a single grain α embedded in ϵ° , (17) is a simple sum of the average contribution of each grain. From elementary electrostatics, when an isotropic dielectric sphere is placed in a uniform electric field E° , the sphere is polarized; t^{α} is connected with this polarization. Equation (14) sums individual "dipoles" embedded in ϵ° and does not account for the fact that any given dipole sees a medium in which other "dipoles" are present. The factor $(I + \langle GT \rangle)^{-1}$ in (13) is the Lorentz correction which accounts for the presence and the polarization of other grains by replacing these grains by a uniformly polarized medium. The important point is that in (16) the grain α is embedded not in ϵ° , as in (14), but in a uniformly "polarized" medium; however, interactions between grains are still neglected.

The approximation (14) has been used in many different contexts usually with $\epsilon^{\circ} = \langle \epsilon \rangle$, for example, in electrical conductivity problems by Maxwell[1], (frequency dependent) dielectric problems by Maxwell-Garnett[2,3], thermal conductivity problem by deVries[4], and in solid state physics by Elliott and Taylor[5]. For elastic problems, Kröner[6] used (17). In solid state physics, (14) is called the Average T-matrix Approximation (ATA).

A commonly used approximation is a self-consistent effective medium approximation. "Self-consistency" is a term used in different contexts, often incompatibly. In the present context, the self-consistency means the following: T in (13) depends on ϵ° through $\delta\epsilon$ and G . If ϵ° were chosen so that $\langle T \rangle = 0$, then $\epsilon^* = \epsilon^{\circ}$.

T , let alone $\langle T \rangle$, is not a quantity one expects to evaluate exactly. Self-consistent solutions, however, can be sought for approximations to T . If, for example,

$$T \approx \sum_{\alpha} t^{\alpha} \quad (18a)$$

and

$$\langle T \rangle = 0 \quad (18b)$$

then for the standard problem of isotropic $\delta\epsilon^\alpha$ and spherical grains one finds the following well-known self-consistent effective medium approximation

$$\sum_j f_j \frac{\epsilon^* - \epsilon_j}{\epsilon^* + 2\epsilon_j} = 0 \quad (19)$$

The self-consistency condition is significant. Both the ATA and self-consistent approximation are based on a perturbation series represented by

$$T \approx \sum_{\alpha} t^{\alpha}$$

i.e. the embedding of single grains in a homogeneous medium. Equation (16), however, does not equal (19). The difference is the ATA stops with the above and thereby ignores interactions between grains, while the self-consistent approximation through the additional condition (18b) incorporates interacting grains in an average manner.

The details of how (18b) incorporates some of the interactions requires more analysis than space permits. Physically, one considers a single grain α in a uniformly polarized medium that represents the presence of the other grains. This polarized medium polarizes α , but the polarization of α changes the polarization of the uniform medium, the change in this polarization changes the polarization of α , etc. until the polarization of α and the uniform medium representing the other grains are consistent on the average.

Self-consistent approximations, as defined in the present context, have been used by various investigators, for example, Bruggeman[7] and Landauer[8] for electrical media, Hershey[9], Hill[10], and Budiansky[11] for elastic media. In solid state physics this approximation was derived by Taylor[12] and by Soven[13] and is called the (single site) Coherent Potential Approximation (CPA).

REMARKS

From (10) corrections to ϵ^* beyond the ATA are easily seen to be second order in t . Corrections to ϵ^* beyond the CPA are not as obvious, but are known to be fourth order in t , which implies that the ATA is an approximation to the CPA. The important point is that analysis natural to the scattering theory with infinite order perturbation summation approach can connect condition (18) to a well-defined perturbation analysis and identify the next order corrections. Since the self-consistency condition is a formal expression of intuitive and physical conditions stated for a variety of related problems, the ad hoc flavor of many self-consistent effective medium approximations is placed on clearly defined theoretical grounds by their relation to an explicit perturbation series that has immediately identifiable corrections.

On physical grounds the approximation,

$$T \approx \sum_{\alpha} t^{\alpha}$$

because it neglects interactions between grains and clustering effects is expected to work for well-separated grains of different species embedded in ϵ^0 . For two-phase systems, this means small concentrations of one species hosted in the other. Because the CPA is also based on this same approximation to T , it too is often regarded as a small concentration approximation. This is not completely correct since comparisons with experiment suggest otherwise. Apparently, for many problems the interactions between grains and such clustering effects as "touching" grains are on the average not as important as one would a priori suppose.

For a two-phase medium the CPA is exact for the low concentration limit for either species. One interpretation of the CPA is that of an interpolation formula between these limits, an interpolation performed according to a specific approximation. The interpretation, of course, is not always useful for problems involving porous materials.

Another important point about the CPA is that it is the best possible approximation using statistical information about single grains, e.g. volume fraction, texture, etc. It is the best possible in the sense that it evaluates every perturbation term that can be evaluated with only single grain distribution functions. Corrections to the CPA require knowledge of two-grain distribution functions.

There is more than one way to relate effective medium approximations to perturbation theory. Elliot et al.[14] and Yonezawa and Morigaki[15] review these ways for solid state physics problems. In a series of papers[16-23] and with a specific approach, Hori and Yonezawa do an extensive analysis of electrical, thermal, and magnetic problems. Gubernatis and Krumhansl[24,25] discuss the analysis as applied to elastic problems. Other references[26-32] treat a variety of problems both from the intuitive and more formal viewpoints. In some cases comparisons with experiment are given. This list is not close to being definitive.

In closing, I emphasize that what I have discussed by example is an approach, scattering theory with infinite order perturbation summation, to problems associated with inhomogeneous media that is formally general, gives a specific recipe for application, and has potential for error analysis.

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